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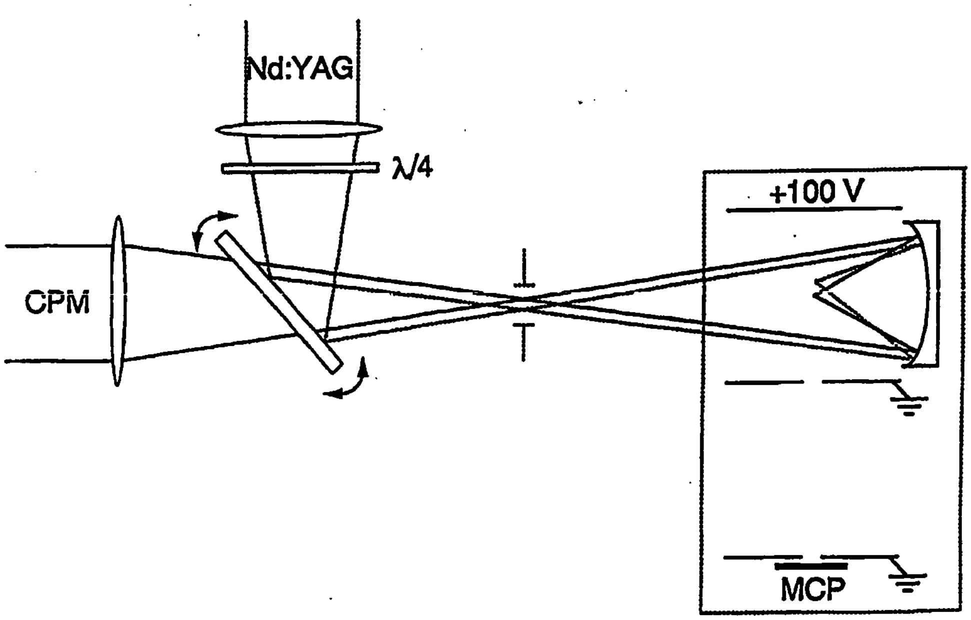

(57) Abstract

In order to manipulate molecules at the molecular level, the molecules are placed in intense, non-resonant laser radiation. The laser radiation is of sufficient intensity to Stark shift the molecular energy levels that the molecules occupy and of sufficient duration and intensity to permit the center-of-mass of said molecules to be spatially manipulated by the gradient of the intensity of the laser radiation in a controllable manner.

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METHOD AND APPARATUS FOR MANIPULATING MOLECULES

This invention relates to a method and apparatus for manipulating molecules at the molecular level.

It is known that focused laser beams from continuous wave lasers apply strong forces to mesoscopic (micron sized) molecules, organelles in biological systems and other mesoscopic particles (see, for example A. Ashkin et. Al., Nature, **348** 346 (1990); S. Chu, Science **253,** 861 (1991)) . The force arises from the intensity gradient of the laser field, which creates a difference in the amount of light diffracted (or reflected) from one side of the particle compared with the other. The imbalance in the momentum imparted leads to a force that is strong enough to direct the mesoscopic particle towards (or away from) the focus of the laser beam, trapping them in (or expelling them from) the region of high light intensity. As the beam is moved about, the trapped molecules move with it. Called optical tweezers, these systems are now widely used in laboratories and are commercially available.

The methods of applying forces to mesoscopic particles do not apply to small molecules since they are not large enough to experience a significant difference in the number of photons impinging on each side.

C. Savage, Aust.J.Phys., **49**, 745 (1996) reviews techniques developed since 1970 (A.Ashkin, Phys. Rev. Lett., **25**, 1321 (1970)) wherein large forces are applied to atoms by exciting them on or near resonance with an atomic transition. In one of the forms of atomic manipulation, the force arises from the preferential adsorption of light for those atoms that are Doppler shifted into resonance. The photons transfer their momentum from the laser beam to the atom during absorption. During spontaneous emission, the photon momentum is given back but since the absorbed photons come only from one direction and the emitted photons go in all directions, there is average

momentum transfer to the atom in direction opposite to the direction of propagation of the laser light.

The methods of applying forces to atoms do not apply to molecules since the spectrum of molecules is so complex that it is not possible to find an isolated resonance on which that resonant force could be applied and because the transition dipole elements between energy levels are typically weak.

An object of the invention is to circumvent these disadvantages of the prior art.

According to the present invention there is provided a method of manipulating molecules at the molecular level, comprising the steps of placing molecules in the gradient of an intense, non-resonant laser radiation, said intensity of said laser radiation being sufficient to Stark shift the molecular energy levels that said molecules occupy, and said laser radiation being of sufficient duration and intensity to permit the center-of-mass of said molecules to be spatially manipulated by the gradient of the intensity of the laser radiation in a controllable manner.

As will be well known by those skilled in the art, the Stark effect causes a shifting of spectral lines in the presence of an electric field. Typically, the intensity and gradient of the laser radiation should be such that the energy imparted to the molecule in the direction of the intensity gradient is not substantially less than the thermal energy of the molecule in that direction and the intensity of the laser radiation should less than the intensity required to substantially ionize or dissociate the molecule.

The molecules may be placed in the gradient of intense, non-resonant, laser radiation, by forming them into a molecular beam traveling substantially perpendicular to the direction of propagation of the said laser radiation, the molecular beam traversing a range of intensity gradients. Alternatively, they may be placed in the gradient of an

intense, non-resonant, laser radiation, by forming a molecular beam traveling substantially parallel to the direction of propagation of said laser radiation so that said molecules are confined in two dimensions while propagating freely in the third direction.

The intensity distribution of said laser radiation may be formed with devices such as lenses, mirrors, axicons, and hollow core fibres. Alternatively, the intensity distribution of said laser radiation may be formed by two or more laser beams so as to form a standing or traveling wave intensity pattern of the laser radiation.

In another embodiment the laser radiation can be in the form of a tapered laser beam and the molecules are funneled to a smaller area as a result of travelling in the direction of the taper or funneled to a larger area as a result of travelling in a direction opposite to the direction of the taper. The tapered laser beam can be produced by means of a tapered hollow core fiber.

If desired the molecular beam, and the gradient of the intensity distribution of said laser radiation can have a form such as to focus or defocus the molecular beam in a controllable manner. Additional strong non-resonant laser beams interacting with the molecules can be provided to further control the properties of said molecular beam, the laser beams creating, in this manner, a compound molecular lens. The focusing, defocusing or collimation of the molecular beam can be manipulated by the compound molecular lens.

The laser radiation can be arranged to create a potential well deeper than the kinetic energy of the molecules such that said molecules are trapped in three dimensions and said laser radiation is moved accelerated or decelerated adiabatically to alter the spatial location of said trapped molecules. After the molecules have been accelerated or decelerated, said laser radiation can be switched off rapidly, in the order of 10^{-13} - 10^{-12} seconds, to produce a field-free molecular beam

moving at substantially constant velocity. The molecules can be focused by the molecular lens so as to be deposited onto a surface over a well defined spatial area of the surface.

The invention can be used to spatially separate the molecules on the basis of their interactions with said laser radiation .

It can also be used to separate molecules according to their polarizability by selectively trapping the most polarizable molecules from a mixture of molecules in thermal equilibrium. The intense non-resonant radiation can also be used to steer the molecules by single or multiple deflections according to their polarizability and mechanical variables, with the radiation creating a straight, curved or tapered waveguide for the molecules.

The inventive method can also be used to apply molecules to a substrate in a defined pattern.

In a typical example, laser manipulation can be used to focus molecules to a region where the density of molecules is greater than the surroundings The effect depends on the energy ratio between the kinetic energy of the molecules and the depth of the laser induced potential well. The kinetic energy is determined by the velocity and the mass, while the well depth depends on the molecular polarizability and the laser intensity. For I_2 molecules at a peak intensity of $10^{11}Wcm^{-2}$ the well depth is $V_w = 2.1$ meV. Assuming a velocity of 500m/sec the energy ratio is $R=E_{kin}/V_w=159$. For an elliptical Gaussian distribution of the laser radiation the molecular beam reaches is highest density at a focal length (f) given by: $\frac{f}{\omega_{0y}} \approx \sqrt{R}\cot\left(\frac{2\omega_{0y}}{\omega_{0z}\sqrt{R}}\right) \approx 9($

The upper limit of the laser radiation that can be used for manipulating molecules is determined by the need to avoid substantial ionization. At far-off resonance

frequencies, the ionization threshold scales approximately as the fourth power of the ionization potential. Different molecules have different ionization potentials. A hydrogen molecule will not substantially ionize in one microsecond up to about $4x10^{13}$ W/cm^2.

The lower limit is determined by the requirement that the energy imparted to the molecule by the laser radiation in the direction of the intensity gradient is not substantially less than the thermal energy of the molecule in the direction of the intensity gradient. The energy imparted to the molecule is determined by the polarizability, the intensity gradient and the duration of the interaction between the molecule and the laser radiation. The polarizability is a molecular property. For a hydrogen molecule, the average polarizability is 0.8 A^3 and for Rb_2 molecules, it is 70 A^3. For many applications the lower limit is typically in the order of 10^9W/cm^2.

The force by which the molecules are manipulated depends on the gradient of the Stark shift. The gradient of the Stark shift is determined by the intensity distribution of the laser beam and the polarizability of the molecule. In a typical example in which an intensity gradient of $2x10^{15}$ Wcm^{-3} is established with a focussed Nd:YAG laser, CS_2 molecules experience a force of approximately $3x10^{-16}$ Newtons resulting in an acceleration of $2.5x10^9$ ms^{-2}.

Typically, the laser beam may be generated with the aid of a Nd:YAG or CO_2 laser, but other lasers capable of producing a sufficiently strong non-resonant radiation could be used.

The invention uses non-resonant radiation. It does not rely on the linear properties of light interactions such as reflection, refraction and diffraction as in the case of optical tweezers, neither does it rely on resonance as in the case when atoms are manipulated by laser light. The invention relies on the nonlinear interaction of the laser beam with

the molecules to allow them to be manipulated spatially in a controllable manner. The invention is applicable to all types of molecules.

According to the present invention, there is also provided an apparatus for manipulating molecules at the molecular level, comprising a cavity for containing molecules; at least one laser for generating intense, non-resonant radiation, the intensity of said radiation being sufficient to Stark shift the molecular energy level that said molecule occupies; and means for controlling the intensity and the duration of said laser radiation so as to permit the center-of-mass of said molecules to be spatially manipulated by the gradient of the intensity of the laser radiation in a controllable manner.

The invention will now be described in more detail, by way of example only, with reference to the accompanying drawings, in which:-

Figure (1a) shows several calculated classical trajectories in the {y,z} plane. A cut through the Stark shift inducing the deflection is shown in Figs. 1b. The intensity distribution of the laser radiation is spherical Gaussian with spot size ω_0. The beam of iodine molecules is moving initially only in the z-direction has an initial velocity of 10^3 $msec^{-1}$. The peak laser intensity is $I_m=5x10^{12}$ Wcm^{-2}. For zero initial angular momentum, the well depth in Fig. (1b) is V_w=120mev, a factor of 12 smaller than the molecular kinetic energy. Figure (1a) shows that the molecular density reaches a maximum at a point y = 0, z = -f referred to below as the focus of the "molecular lens". A cut through the molecular beam at z = -f is given in Fig. (1c), showing a sharp peak at y =0. An alternative means of characterizing the laser lens is in terms of the image size (W) and the distance (D) from the laser axis of the image plane (Fig. (1a)). At z = -D the initial molecular distribution has been focused to with a factor W of its spread.

Figure 2a shows the focal distance f (— - — -), the image size W (——) and the distance of the image from the laser beam D (---) plotted as a function of the energy ratio $R=E_{kin}/V_w$. The calculation was performed for zero initial angular momentum and molecules moving initially only in the z-direction. The inset shows a magnified view of the strong interaction (small R) regime. The intensity distribution of the laser intensity is spherical Gaussian and all distances are measured in units of the laser spot-size ω_0.

Figures 2b and 2c show the focal distance f and the image size W plotted as a function of the energy ratio R for an elliptical Gaussian intensity distribution with different values of the aspect ratio $\eta=\omega_{0y}/\omega_{0z}$ (...... $\eta=1$; - - - - $\eta=2$; ———$\eta=4$; ——— $\eta=7.5$). The circles in Fig. 2b show the analytical approximation of Eq. 1

Figure 3a is a schematic of an experimental apparatus used in one embodiment of a deflection and focusing experiment. A 10 ns pulse from a Nd:YAG laser is focussed by a lens, passes through a quarter wave ($\lambda/4$) plate, and then combined with an independently focussed femtosecond diagnostic pulse from a CPM laser at a beam splitter. Both pulses are then focussed inside the time-of-flight mass spectrometer by a parabolic mirror.

Figure 3 b is a schematic of inside of the time-of-flight mass spectrometer. Electrodes, separated by 3 cm, define the acceleration and drift region of the spectrometer. The arrival time of ions with respect to the time of their ionization by the femtosecond pulse is measured using a micro-channelplate (MCP) detector connected to a digital oscilloscope (not shown). Molecules enter the time-of-flight chamber from a supersonic jet with nozzle diameter of 250 microns. The Nd:YAG and CPM beams (details in Fig 3a) are shown propagating in the x-direction. They intersect the molecular beam 8 cm from the jet nozzle.

Figure 4 is a plot of a portion of three records of arrival times of CS_2 molecular ions. The solid curve shows the arrival time when the Nd:YAG laser beam was not used. Since these molecules are un-deflected, they serve as a reference for the deflected molecules. The dotted curve shows arrival times for molecules that traverse the Nd:YAG beam on the side of the microchannel detector (as illustrated in the right insert). The dashed curve shows arrival times for molecules that that traverse the Nd:YAG beam on the side away from the microchannel detector (as illustrated in the left inset). Changes in arrival times correspond to velocity shift in the y direction as shown in the upper scale.

Figure 5 shows the change in arrival time of the CS_2^+ molecules caused by the deflection beam and the associated change in transverse velocity plotted as a function of the relative position of the focus of the deflecting and the ionizing laser beams. The filled data points were obtained using circularly polarized Nd:YAG light while the open points were obtained with linearly polarized light of the same pulse energy. The derivative of the measured focal distribution (with an arbitrary vertical scale) is also shown by the solid curve.

Figure 6 shows calculated results where a 2 µm diameter beam of I_2 (J=0) molecules is focussed by a two component molecular lens consisting of two laser beams. The first laser beam is at y=z=0, has peak intensity I_1,$=3\times10^{11}$ Wcm^{-2} and focal spot sizes ω_{y1}=2µm, ω_{z1}=1µm. The second laser is at y=0, z=-90 µm, has peak intensity I_2=1×10^{12} Wcm^{-2} and ω_{y2}=1µm, ω_{z2}=2µm. The molecular beam velocity is 500 m/sec.

Figure 7 (inset) shows calculated results where a 2 µm diameter beam of I_2 molecules is focused by means of laser beam, then collimated by means of a second laser beam. The focusing laser beam is of peak intensity 2×10^{11} Wcm^{-2} with ω_{0y}=8 µm and ω_{0z}=2 µm. The collimating laser is of the same peak intensity with ω_{0y}= ω_{0z}=4 µm

and the molecular beam velocity is 500 m/sec. Figure 7 also shows the results of calculations where a third segment of the laser is used to steer the focused and collimated molecular beam in a desired direction. The steering laser is of peak intensity $5x10^{11}$ Wcm^{-2} and the dimensions of the major and minor axes are 8 and 2 μm. The major axis is rotated by 50 degrees with respect to the molecular beam propagation direction and the beam centre is at y=6μm, z=-2600μm

Figure 8 illustrates the possibility of steering a distribution of velocities and impact parameters into a single direction. The molecules enter an elliptical Gaussian intensity distribution at an R-dependent incident angle with respect to the major axis and exit at the same scattering angle. The peak intensity is I_m=$5x10^{12}$ Wcm^{-2}, the laser spot sizes are ω_{0y}=4 μm and ω_{0z}=2 μm and the I_2 (J=0) molecular beam consists of a ±10% velocity distribution centered about 200 m/sec.

Figure 9 shows methods for separating two groups of molecules represented by A and B and differing in their velocity, mass or polarizability. In figure 9a the laser radiation is guided by a curved hollow core wave-guide. Only those molecules which experience a laser-induced acceleration equal to or exceeding the centripetal acceleration given by v^2/r (where r is the radius of the curved wave guide and ν is the tangential velocity of the molecule). can remain within the high intensity region of the laser. All other molecules will collide with the walls of the wave-guide and hence remain inside of the wave guide. In figure 9b one capitalizes on the fact that the intense field associated with the laser radiation can be configured so as to sequentially deflect out of a molecular beam the most strongly interacting component(s)., The strength of the interaction being determined by the mass, velocity or polarizability of the component(s).

Theoretical Basis

In an electric field gradient the molecules seek the region of highest intensity, where their energy is minimized. An intensity gradient is found naturally in any laser beam. For example, a Gaussian intensity distribution is given by $I(x,y,z)=I_m(x)\exp-(y^2/\omega_{0y}{}^2+z^2/\omega_{0z}{}^2)$. It follows that a laser field can serve to manipulate (focus, steer, reflect, waveguide and trap) the center-of-mass motion of the molecules.

In the far-off resonance limit, assuming a cylindrical laser beam, the complete Hamiltonian is given as

$$H=H_{rel}+1/2m(d^2/dy^2+d^2/dz^2)-\mu_{ind}(y,z;t).\varepsilon\,(y,z;t) \quad (2)$$

where H_{rel} is the field-free Hamiltonian for the relative (internal) motion and the induced dipole is $\mu_{ind}(y,z;t)=\alpha.\varepsilon\,(y,z;t)$ where α is the dynamic polarizability tensor. Manipulation of the centre of mass motion arises from the spatial dependence of the laser radiation. The qualitative difference between the Hamiltonian (Eq. 2) and the familiar Hamiltonian [1] used in theories of atomic focusing arises from the anisotropy of typical molecular polarizabilities and from the fact that a 2-level description cannot be applied to molecules.

Equation (2) shows that the laser field can also align nonspherical molecules due to the angular dependence of $\alpha.\varepsilon^2$ [2] While rotational motion is induced by a gradient of the molecular size-scale (several Angstroms) translational motion is induced by a gradient of the laser-beam dimension (several μm). The consequent large time-scale disparity between the rotational and center-of-mass motions and the long pulses required, allows adiabatic separation n of the rotational and the translational modes.

First a quantum mechanical solution is found for the rotational motion at fixed values of the field amplitude ε and next the resulting eigenvalues are used as effective potentials for the center-of-mass motion. The center-of-mass translation is generally

adequately described by classical mechanics.[3-5]

Hamilton's equations for the center-of-mass motion are propagated on the numerically calculated space dependent potentials evolving on the parametrically time and space dependent potentials $E^{JMK}[\varepsilon(y,z;t)]$ where J, M and K are determined by the mode of preparation of the system. It is also useful to note that to zero order the depth of the laser-induced effective well is $V_w \cong \alpha_{\parallel} \varepsilon_m^2 / 4$ where $\alpha_{\parallel}$ is the component of the polarizability along the molecular axis.

Figure (1a) shows several calculated classical trajectories in the {y,z} plane. A cut through the Stark shift inducing the deflection is shown in Figs. 1b. The intensity distribution of the laser radiation is spherical Gaussian with spot size ω_0. The beam of iodine molecules with diameter $2\omega_0$ moving initially only in the z-direction has an initial velocity of 10^3 $msec^{-1}$. The peak laser intensity is $I_m=5x10^{12}$ Wcm^{-2}. For zero initial angular momentum, the well depth is V_w=120mev, a factor of 12 smaller than the molecular kinetic energy. Figure (1a) shows that the molecular density reaches a maximum at a point y = 0, z = -f referred to below as the focus of the "molecular lens". A cut through the molecular beam at z = -f is given in Fig. (1c), showing a sharp peak at y =0. An alternative means of characterizing the laser lens is in terms of the image size (W) and the distance (D) from the laser axis of the image plane (Fig. (1a)). At z = -D the initial molecular distribution has been focused to with a factor W of its spread.

While f is independent of the initial beam diameter, W is a function of the ratio the diameter of the molecular and optical beams. For the conditions in figure 1 the radius of the region where the molecular density exceeds 1/e of its value at y=0 is W_M = 0.03 ω_0. W_M provides a measure of the resolution of the molecular lens .

As the ratio $R = E_{kin}$,/V_w. between the kinetic energy and the well depth decreases, both the focal length *f*, and the width and distance parameters decrease until *W* shrinks to a point and D = f. This behaviour is illustrated quantitatively in figure 2a which shows the focal distance *f* (— - — -), the image size *W* (——) and the distance of the image from the laser beam *D* (---) plotted as a function of the energy ratio $R=E_{kin}/V_w$. The calculation was performed for zero initial angular momentum and molecules moving initially only in the z-direction. The inset shows a magnified view of the strong interaction (small R) regime. The intensity distribution of the laser intensity is spherical Gaussian and all distances are measured in units of the laser spot-size ω_0. To provide a measure of the laser and molecular beam parameters involved, we note that R=5, $W/\omega_0=0.2$ corresponds to a to maximum intensity of $I_m= 10^{12}$ Wcm^{-2} with initial velocity v=300m/sec. The same point in figure 2 would be obtained for nv and n^2I_m for any value of n.

Figures 2b and 2c show the focal distance f and the image size W plotted as a function of the energy ratio R for an elliptical Gaussian intensity distribution with different values of the aspect ratio $\eta=\omega_{0y}/\omega_{0z}$ (...... $\eta=1$; - - - - $\eta=2$; ————$\eta=4$; ——— $\eta=7.5$). The circles in Fig. 2b show the analytical approximation of Eq. 1

Both *f* and *D* are linear in the energy ratio while the image size *W* has a fast turn-on and reaches a nearly flat plateau.

Experimental Basis:

A 10 ns Nd:YAG laser with maximum laser energy of 300 mJ is used to generate an intense polarized, laser beam to focus and deflect the molecules. As shown in Figure 3a , the laser beam is focussed by a 1m focal length lens and then passes through a quarter wave ($\lambda/4$) plate, and is combined with an independently focussed femtosecond diagnostic pulse at a dichroic beam splitter.

The diagnostic beam has a duration of 80 fs, a wavelength of 625 nm and a maximum energy of 200 μJ. The 625 nm pulse is produced by amplifying one output pulse from a colliding-pulse mode-locked dye laser in a five-stage dye amplifier pumped with the 532 nm 10 Hz pulse from the same Nd:YAG laser used for deflection and focusing.

The focus of both beams are measured and independently positioned at a focal spot 1 meter in front of the target chamber. The focal spot is imaged into the chamber with a demagnification of 20:1 by a 5 cm focal length 2.5 cm diameter parabolic mirror. The focal spot inside the chamber is $\omega_0 = 7$ microns for the Nd:YAG beam and $\omega_0 = 2.5$ microns for the femtosecond diagnostic beam. For measurement, the femtosecond pulse is focused to a position just below the Nd:YAG focal spot position and arrived at a delay time just sufficient to allow the molecules to move from the Nd:YAG focus to the diagnostic region.

The target chamber shown schematically in Figure 3b is fitted with a supersonic CS_2 molecular beam with axis perpendicular to the axis of the laser beam. For some experiments the CS_2 molecules are buffered with neon to produce rotationally cold molecules. The vacuum chamber has a base pressure of 10^{-8} mBarr and rises to an average pressure of 10^{-7} mBarr when operated with a neon buffer at 10 Hz.

The time-of flight chamber with the flight axis perpendicular to both the beam axis and the direction of propagation of the laser beam, consisted of three 4 cm diameter electrodes forming the acceleration and drift regions of the time-of-flight mass spectrometer. A 100 V potential provides a 33 V/cm acceleration field. .

Measurement of molecular deflection consists of producing singly charged molecular ions by multiphoton ionization. Multiphoton ionization ensures that the measured

molecules are produced very near the focal position of the femtosecond pulse. After formation by multiphoton ionization, the ions are accelerated towards the ground electrode passed through a small hole in the electrode and entered a 3 cm field free drift region defined by a third electrode maintained at ground. The ions that pass through a hole in the final electrode and are detected on a microchannel plate detector where their arrival time with respect to the femtosecond pulse is recorded on a digital oscilloscope. Molecules with an initial velocity in the time-of-flight direction and moving towards the detector arrive earlier than those with a velocity away from the detector.

By measuring the arrival time of the singly charged molecular ions with and without the Nd:YAG beam, we determine the magnitude of the molecular deflection. By scanning the femtosecond beam with respect to the Nd:YAG focus, we observe how molecular deflection depends on the position in the Nd:YAG beam.Figure (4) is a portion of three time-of-flight records for CS_2^+. Each curve is the average of 1000 laser shots and the laser intensity is 10^{12} Wcm^{-2}. The solid curve shows the arrival time when the Nd:YAG laser beam is not used. Since these molecules are undeflected, they serve as a reference for the deflected molecules. The dotted curve shows arrival times for CS_2^+ produced from those molecules that traverse the Nd:YAG beam at a distance of approximately $\omega_0/2$ from beam centre displaced towards the microchannel detector. The arrival time is delayed indicating that the molecules are deflected away from the detector. The dashed curve shows arrival times for molecules that that traverse the Nd:YAG beam at a distance of approximately $\omega_0/2$ from beam centre displaced away from the microchannel detector. The arrival time is advanced compared with the undeflected molecules (solid curve) indicating that the molecules are deflected towards the detector. The geometry is illustrated by insets in the figure.

Figure 5 shows the change in arrival time (with respect to the arrival time of

undeflected molecules) of the CS_2^+ caused by the deflection of the CS_2 molecules and the associated change in transverse velocity plotted as a function of the relative position of the focus of the deflecting and the ionizing laser beams. The shifts are measured at the centre position of the half-maximum of the time-of-flight spectra (similar to those in Figure 4). The filled data points were obtained using circularly polarized Nd:YAG light with peak intensity of 10^{12} Wcm^{-2} while the open points were obtained with linearly polarized light of the same pulse energy. The derivative of the measured focal distribution (with an arbitrary vertical scale) is shown by the solid curve.

The results shown in figures 4 and 5 demonstrate that CS_2 molecules experience a force of approximately $3x10^{-16}$ Newtons resulting in an acceleration of $2.5x10^9$ ms^{-2}. This implies a Stark shift of approximately 7 meV. Since a multi-mode pulse was used, those skilled in the art will understand that significantly deeper potential wells can be created.

This experiment has been repeated with I_2 molecules and we have observed deflection by a Nd:YAG beam yielding potential well depths of about 6.5 meV for I_2. The experiment has also been repeated using a CO_2 laser beam. We have measured a potential well depths of 4 meV at a peak intensity of $4x10^{11}$ Wcm^{-2}, the maximum intensity that our CO_2 laser could produce.

Those skilled in the art will understand that any laser can be used to deflect molecules provided it reaches a high enough intensity that the Stark shift is significant compared to the thermal energy of the molecule. They will also understand that Stark shifts with magnitude similar to those measured for iodine and CS_2, can be achieved for other molecules including such molecules as H_2.

Applications

There are numerous important practical application of the present invention.

The molecules can be confined in two dimensions, with the molecules propagating substantially freely in the third dimension, thereby creating a wave-guide effect. Examples of ways to produce the laser beam that is responsible for guiding molecules include the use of spherical lenses or mirrors, cylindrical lenses or mirrors, axicons or hollow core fibres.

A tapered laser beam can concentrate the molecules in two dimensions with the molecules propagating substantially freely in the third dimension, producing a funneling effect. The optimum effect will occur if the taper of the funnel is gradual enough so that the molecules do not exchange energy between the confined and non-confined direction. Tapered hollow core fibres can be used for this purpose, but other focusing methods are possible.

Figure 6 shows how two intense laser beams can be combined to reduce significantly the image size compared to a single molecular lens. In the example shown in figure 6 we chose the initial I_2 (J=0) molecular beam diameter to be 2 μm with initial velocity of 500 m/sec in the z-direction. The first laser beam is at y=z=0 and has peak intensity I_1,$=3\times10^{11}$ Wcm^{-2} and spot size ω_{0y}=2μm, ω_{0z}=1μm. The second laser is at y=0, z=-90 μm and has peak intensity $I_2=1\times10^{12}$ Wcm^{-2} and spot size ω_{0y}=1μm, ω_{0z}=2μm..

A laser beam can be used to collimate/focus/expand a neutral molecular beam. For instance, a beam of laser-focused molecules which otherwise diverges after exiting the focusing laser beam could be collimated by means of a second laser as shown in Figure(7(inset)). The beam can remain collimated over distances of millimeters. The precise length and the waist of the collimated molecular beam can be varied by changing the molecular beam velocity, the intensity and the distance between the focusing-and collimating laser beams. Figure 7 also shows that the addition of a third

laser beam can be used to steer the focussed and collimated molecular beam.

Molecules can be steered into substantially the same direction, even when they have a substantial velocity spread in their direction of propagation and even when they have a substantial range of impact parameters. This is illustrated by theoretical calculations one example of which is shown in figure 8. In this case molecules enter an elliptical Gaussian intensity distribution at an R-dependent incident angle with respect to the major axis and exit at the same scattering angle. In the example shown in figure 8 the peak intensity is $I_m=5\times10^{12}$ Wcm $^{-2}$, the laser spot sizes are $\omega_{0y}=4$ μm and $\omega_{0z}=2$ μm and the I_2 (J=0) molecular beam consists of a ±10% velocity distribution centered about 200 m/sec. Theoretical calculations show that this is an important special case of beam steering where a distribution of velocities and impact parameters is steered into essentially a single direction. The angle between the incident and scattered molecular beams is controlled by the energy ratio $R=E_{kin}/V_w$. As R increases this angle increases until, in the limit of collinear approach, the laser beam serves again as a focusing lens.One of the applications of such a device is to produce a short-pulsed molecular beam. The molecular density in the scattered direction is fully controlled by the laser pulse duration, achieving substantially better time-resolution than with conventional (mechanically) pulsed molecular beams.

The acceleration of molecules by laser radiation that allows us to deflect, focus or steer molecules can be used to separate groups of molecules, differing in their velocity, mass or polarizability. Traditionally it has been the frequency specificity of lasers which has been exploited in laser separation processes. There are two economic limitations to this approach. Firstly, since no single class of lasers covers the entire spectrum, for any given process a specific laser is needed and/or developed. The most notable case in point is the copper vapor laser pumped dye systems used in Uranium enrichment plants (see for example R.P. Hackel and B.E. Warner, Proceedings-of-the-SPIE The International Society for Optical Engineering ,**1859**, 120-9,1993) Secondly,

stringent frequency stability is usually imposed on the laser. The inevitable consequence of this is a reduction in the economic efficiency of the laser. In contrast, the mechanism which is used by this invention to manipulate molecules is a non resonance process. Any sufficiently intense non-resonant laser radiation can be used, and the laser can be operated in its most energy efficient mode. Trace impurity removal or isotope separation are but two such examples in which these properties may be used.

Figure 9 shows schematics of two possible approaches to achieve separation. In the example shown in figure (9a), the intense laser beam is guided by a curved hollow-core optical wave guide. Molecules entering the wave guide traveling parallel to the laser beam propagation direction are selectively guided. Only those molecules which experience a laser-induced acceleration equal to or exceeding the centripetal acceleration given by v^2/r (where r is the radius of the curved wave guide and v is the tangential velocity of the molecule) can remain within the high intensity region of the laser. All other molecules will collide with the walls of the wave guide and hence remain inside of the wave guide.

In the example shown in figure 9b one capitalizes on the fact that the intense field associated with the laser radiation can be configured so as to sequentially filter out of a molecular beam the most strongly interacting component. The strength of the interaction being determined by the mass, velocity or polarizability of the component. A suitable place "collector" can now be used to collect the selected component.

The interrelationship between the properties of the molecular beam and the intense laser radiation can be also used to accelerate or decelerate the molecular beam without altering the trajectory of the molecular beam. The use of a moving high intensity region with that region moving at the same velocity as the molecules in a molecular beam can be used to accomplish this effect. This high intensity region could be the

focus of a laser beam, it could be the intensity of a laser beam contained within a hollow core wave-guide, or it could be the anti-nodes of a standing wave interference pattern. Movement of this region could be accomplished by chirping the laser radiation or by moving the optical components which produce the high intensity region at a fixed or a variable velocity. The potential well depth associated with this high intensity region should be deep enough to confine the molecules to the moving high intensity regions. These molecules are then either accelerated or decelerated by adiabatically increasing or decreasing the speed of motion of the high intensity region.

One of many practical applications of this particular form of molecular manipulation would be to control the impact velocity of a molecule as it strikes a surface. For example, if such a moving intensity region is placed in the collimated beam shown in figure 7 inset, this combined intensity structure could form the basis of a controlled surface deposition technique. An appealing extension is the possibility of using this deposition method to generate novel materials or to enhance the properties of a known material. For example, the ability to steer and guide molecules onto a surface and to concentrate such molecules in ordered arrays in a small regions can be applied to the deposition of nanostructures. Taken together with the dependency of magnetic and electric properties on the degree of order, new materials with pre-designed electric and/or magnetic properties may be produced by this deposition technique

A rapidly moving intensity structure could be combined with tight focus (see, e.g, Fig. 2c or Fig. 6) to produce the molecular equivalent of the focused ion beam (FIB), i.e. a focused molecular beam (FMB). The moving intensity structure could be used to accelerate the molecules towards the focus in an analogues manner to the way an electric field is used to accelerate the ions of a FIB. An important advantage which such a FMB would have over a FIB is that it could be used on insulators and conductors with equal effectiveness since surface charging, a major concern with the

use of a FIB on insulators, is eliminated.

Another appealing application of laser-manipulation of molecules is nanoscale etching of semiconductor wafers. The most common method for etching such wafers involves the following steps:- (1) coating the substrate with a photoresist, (2) projecting a pattern onto the surface this photo resist by means of an optical projection system and a mask, (3) developing the photo resist, (4) etching away the material not protected by the photo-resist by some form of ion sputtering and (5) finally removing the remaining photoresist. Feature size definition using optical projection techniques is limited by diffraction and by the damage associated with ion sputtering. These two limitations impose a fundamental lower bound on further device miniaturization. Manipulation of atoms by optical radiation offers one way of circumventing the above impediment to future miniaturization. Near-resonance, weak-field standing-wave optical techniques have been used for focusing atoms in etching and deposition processes. The technique is capable of producing ca 80 nm wide strips over substantial areas (> 3 x $10mm^2$) . However, it requires an extremely well-collimated (hence ultra-old) atomic beam, it is limited to atoms of simple level structure and to depositing equally-spaced parallel strips with the spacing fixed by the light frequency. In addition, the achievable contrast is limited by the sine form of the gradient which induces focusing and the generality is constrained by the proximity of the laser beam to the surface.

The present invention offers a complementary optical solution to the problem of future miniaturization to that offered by atomic weak-field techniques. The following (2) steps constitute one such approach. (a) An intense laser beam is used to deposit the beam of molecules onto a surface in a controlled manner and with spatial dimensions much smaller than that associated with the beam waist of the laser, as described above. The molecules are chosen to alter the chemical nature of the surface at, and only at the region of the surface covered by such molecules. (b) An auxiliary

method, this could be an additional laser beam or a focused molecular beam, is then be used to remove the products of the surface reaction. For instance, focusing iodine molecules onto a silicon surface followed by laser desorption of Si-I would etch nanoscale features into a silicon substrate.

In the summary, the present invention is believed to have very important ramifications in the field of molecular physics since for the first time it provides a method of carrying out the hitherto impossible task of manipulating molecules in a beam.

Claims:

1. A method of manipulating molecules at the molecular level characterized in that the molecules are placed in the gradient of intense, non-resonant, laser radiation, said intensity of said laser radiation is sufficient to Stark shift the molecular energy level that said molecules occupy, and said laser radiation is of sufficient duration and intensity to permit the center-of-mass of said molecules to be spatially manipulated by the gradient of the intensity of the laser radiation in a controllable manner.

2. A method as claimed in claim 1, characterized in that the intensity and gradient of the laser radiation are such that the energy imparted to the molecule in the direction of the intensity gradient is not substantially less than the thermal energy of the molecule in said direction.

3. A method as claimed in claim 1 or 2, characterized in that the intensity of the laser radiation is less than the intensity required to substantially ionize or dissociate the molecule.

4. A method as claimed in any one of claims 1 to 3, characterized in that the molecules are placed in said gradient of intense, non-resonant, laser radiation, by forming them into a molecular beam traveling substantially perpendicular to the direction of propagation of the said laser radiation, said molecular beam traversing a range of intensity gradients.

5. A method as claimed in any one of claims 1 to 3, characterized in that the method of placing molecules in the gradient of an intense, non-resonant, laser radiation, consists of forming a molecular beam traveling substantially parallel to the direction of propagation of said laser radiation so that said molecules are confined in two dimensions while propagating freely in the third direction.

6. A method as claimed in any one of claims 1 to 5, characterized in that the intensity distribution of said laser radiation is formed with devices selected from the group consisting of: lenses, mirrors, axicons, and hollow core fibres.

7. A method as claimed in any one of claims 1 to 3, characterized in that the intensity distribution of said laser radiation is formed by two or more laser beams so as to form a standing or traveling wave intensity pattern of the laser radiation.

8. A method as claimed in claim 5, characterized in that said laser radiation is in the form of a tapered laser beam and said molecules are funneled to a smaller area as a result of travelling in the direction of the taper or funneled to a larger area as a result of travelling in a direction opposite to the direction of the taper.

9. A method as claimed in claim 8, characterized in that said tapered laser beam is produced by means of a tapered hollow core fiber.

10. A method as claimed in claim 1, characterized in that the molecules are placed in the gradient of intense, non-resonant, laser radiation, by forming them into a molecular beam, and the gradient of the intensity distribution of said laser radiation having a form such as to focus or defocus said molecular beam in a controllable manner.

11. A method as claimed in claim 10, characterized in that additional strong non-resonant laser beams interacting with the molecules are provided to further control the properties of said molecular beam, said laser beams creating, in this manner, a compound molecular lens.

12. A method as claimed in claim 11, characterized in that the focusing, defocusing or collimation of said molecular beam is manipulated by said compound molecular lens.

13. A method as claimed in claim 11, characterized in that the gradient of the intensity distribution of said laser radiation has such a form as to steer said molecular beam in substantially one direction in a controllable manner.

14. A method as claimed in claim 1, characterized in that said laser radiation creates a potential well deeper than the kinetic energy of said molecules such that said molecules are trapped in three dimensions and said laser radiation is moved adiabatically to alter the spatial location of said trapped molecules.

15. A method as claimed in claim 14, characterized in that said laser radiation is accelerated or decelerated adiabatically to accelerate or decelerate the molecules in the beam.

16. A method as claimed in claim 15, characterized in that, after said molecules have been accelerated or decelerated, said laser radiation is switched off to produce a field-free molecular beam moving at substantially constant velocity.

17. A method of manipulating molecules onto a surface in ordered structures comprising the steps of placing molecules in the gradient of intense, non-resonant, laser radiation, said intensity of said laser radiation being sufficient to Stark shift the molecular energy level that said molecule occupies and said laser radiation being of sufficient duration to permit the center-of-mass of said molecules to be spatially manipulated by the gradient of the intensity of the laser radiation in a controllable manner.

18. A method as claimed in claim 17, characterized in that the energy imparted to the molecule by the laser radiation in the direction of the intensity gradient is not substantially less than the thermal energy of the molecule in the direction of the intensity gradient.

19. A method as claimed in claim 17, characterized in that the intensity of the laser radiation is less than the intensity required to substantially ionize or dissociate the molecules.

20. A method as claimed in claim 17, characterized in that said molecules are focused by a molecular lens so as to deposit said molecules onto a surface over a well defined spatial area of the surface.

21. A method as claimed in claim 20, characterized in that the gradient of the intensity distribution of said laser radiation has a form such as to focus or defocus said molecular beam in a controllable manner and thereby form said molecular lens.

22. A method as claimed in claim 20, characterized in that additional strong non-resonant

laser beams interacting with the molecules are provided to further control the properties of said molecular beam and thereby form said molecular lens as a compound lens.

23. A method as claimed in any one of claims 20 to 22, characterized in that said molecules are deposited onto a surface over a spatial area smaller than the wavelength of the intense, non-resonant, laser radiation..

24. A method of separating one or more species of molecules from a mixture of molecules according to their polarizability or mechanical variables, characterized in that the molecules are placed in the gradient of intense, non-resonant, laser radiation, said intensity of said laser radiation is sufficient to Stark shift the molecular energy level that said molecule occupies, and said laser radiation is of sufficient duration to permit the center-of-mass of said molecules to be spatially manipulated by the gradient of the intensity of the laser radiation in a controllable manner such that the molecules are spatially separated on the basis of their interactions with said laser radiation .

25. A method of separating molecules as claimed in claim 24 according to their polarizability by selectively trapping the most polarizable molecules from a mixture of molecules in thermal equilibrium.

26. A method of separating molecules as claimed in claim 24, characterized in that said intense non-resonant radiation steers said molecules by single or multiple deflections according to their polarizability and mechanical variables.

27. A method of separating molecules as claimed in claim 26, characterized in that said laser radiation creates a straight, curved or tapered waveguide for said molecules.

28. A method of separating molecules as claimed in claim 24, characterized in that said intense non-resonant radiation creates a potential well deeper than the kinetic energy of at least one component of said molecular mixture such that said molecules are trapped in three dimensions and said potential wells are then accelerated or decelerated, and different components of the molecular mixture escape from the

potential wells at different accelerations of said potential wells.

29. A method of making an article which includes at least one operation that requires molecules to be applied to a substrate in a defined pattern, characterized in that molecules are placed in the gradient of intense, non-resonant, laser radiation, said intensity of said laser radiation being sufficient to Stark shift the molecular energy level that said molecule occupies, and said laser radiation is of sufficient duration to permit the center-of-mass of said molecules to be spatially manipulated by the gradient of the intensity of the laser radiation in a controllable manner so that they are applied to said substrate in the defined pattern.

30. A method as claimed in claim 29,characterized in that the energy imparted to the molecules by said laser radiation in the direction of the intensity gradient is not substantially less than the thermal energy of the molecule in the direction of the intensity gradient.

31. A method as claimed in claim 29, characterized in that the intensity of said laser radiation is less than the intensity required to substantially ionize or dissociate said molecule.

32. An apparatus for manipulating molecules at the molecular level, characterized in that it comprises:

a cavity for containing molecules;

at least one laser for generating intense, non-resonant radiation, the intensity of said radiation being sufficient to Stark shift the molecular energy level that said molecule occupies; and

means for controlling the intensity and the duration of said laser radiation so as to permit the center-of-mass of said molecules to be spatially manipulated by the gradient of the intensity of the laser radiation in a controllable manner.

33. An apparatus as claimed in claim 32, characterized in that it further comprises means for forming said molecules into a molecular beam in said cavity, thereby permiting the center-of-mass of said molecules in said beam to be manipulated in a

controllable manner.

34. An apparatus as claimed in claim 32, characterized in that the energy imparted to the molecules by the laser radiation in the direction of the intensity gradient is not substantially less than the thermal energy of the molecule in the direction of the intensity gradient.

35. An apparatus as claimed in any one of claims 32 to 34, characterized in that said laser is selected the group consisting of: Nd:YAG and CO_2.

36. An apparatus as claimed in claim 33, characterized in that said cavity includes a support for a substrate, and said molecular beam directable onto said substrate.

37. A method of manipulating molecules at the molecular level, characterized in that molecules are aligned in intense, laser radiation, and said laser radiation is then rapidly switched off to produce free-field aligned molecules.

38. A method as claimed in claim 37, characterized in that said field is switched off in a time faster than molecular rotation, which is in the order of 10^{-13} - 10^{-12}.

39. A method as claimed in claim 38, characterized in that the direction of polarization is rotated in space with adiabatically increased speed to create spinning molecules.

40. An apparatus for manipulating molecules at the molecular level, characterized in that it comprises:

a cavity for containing molecules;

at least one or more lasers for generating intense radiation having an intensity sufficient to Stark shift the molecular energy level that said molecule occupies;

means for controlling the intensity and the duration of said laser radiation said intensity of said laser radiation to permit a molecule with anisotropic polarizability to align with dipole moment of said molecule aligned to the polarization vector of said laser; and

means for rapidly switching off said laser radiation, thereby producing field

free aligned molecules.

41. An apparatus as claimed in claim 40, characterized in that means are provided for forming the molecules into a molecular beam in said cavity.

42. An apparatus as claimed in claim 40, characterized in that said laser radiation is produced by a CO_2 laser, and said means for rapidly switching off said laser radiation comprises a semiconductor normally transparent to said radiation and which is switchably controlled by a laser producing femtosecond above-bandgap radiation.

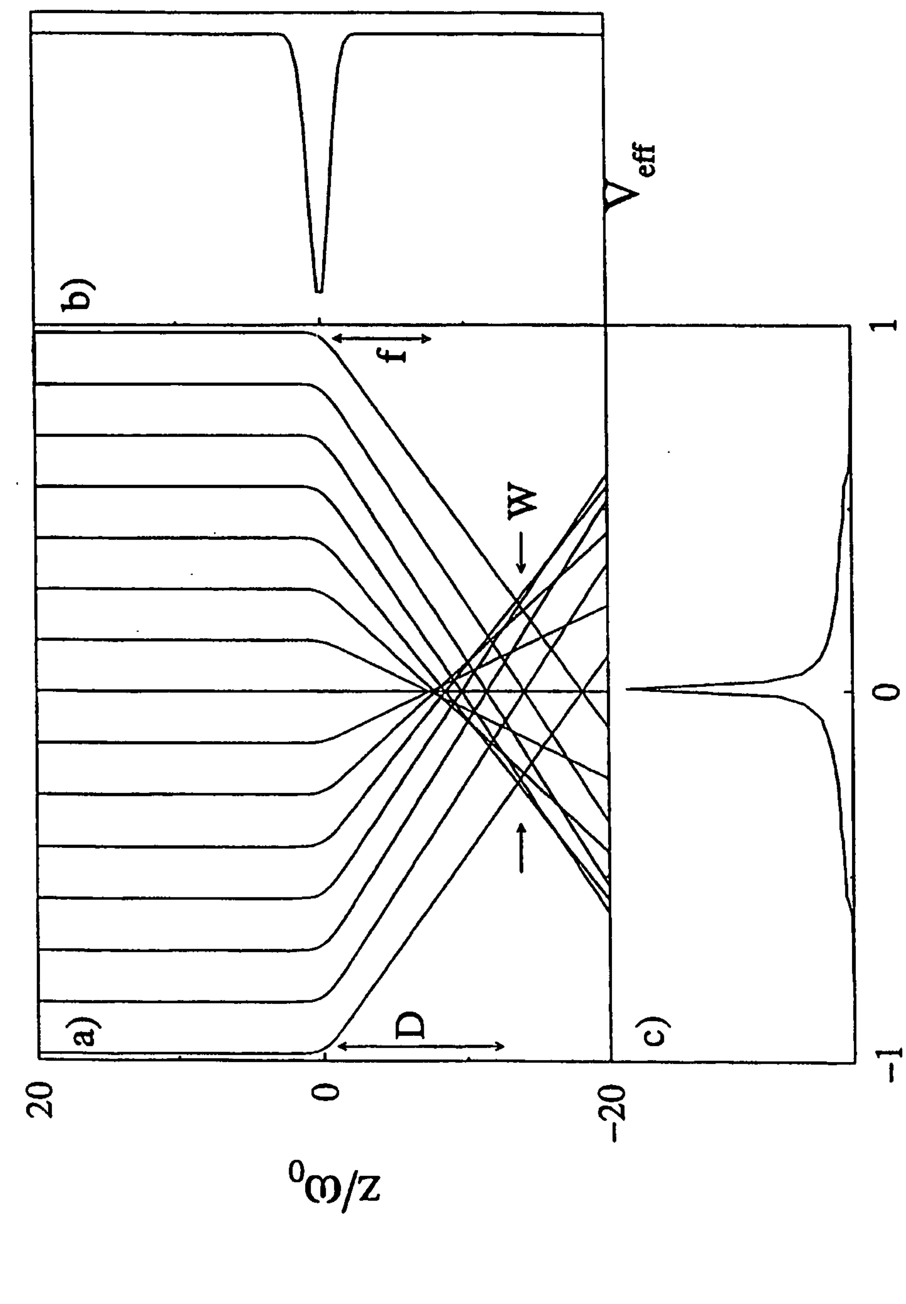


Figure 1

2/13

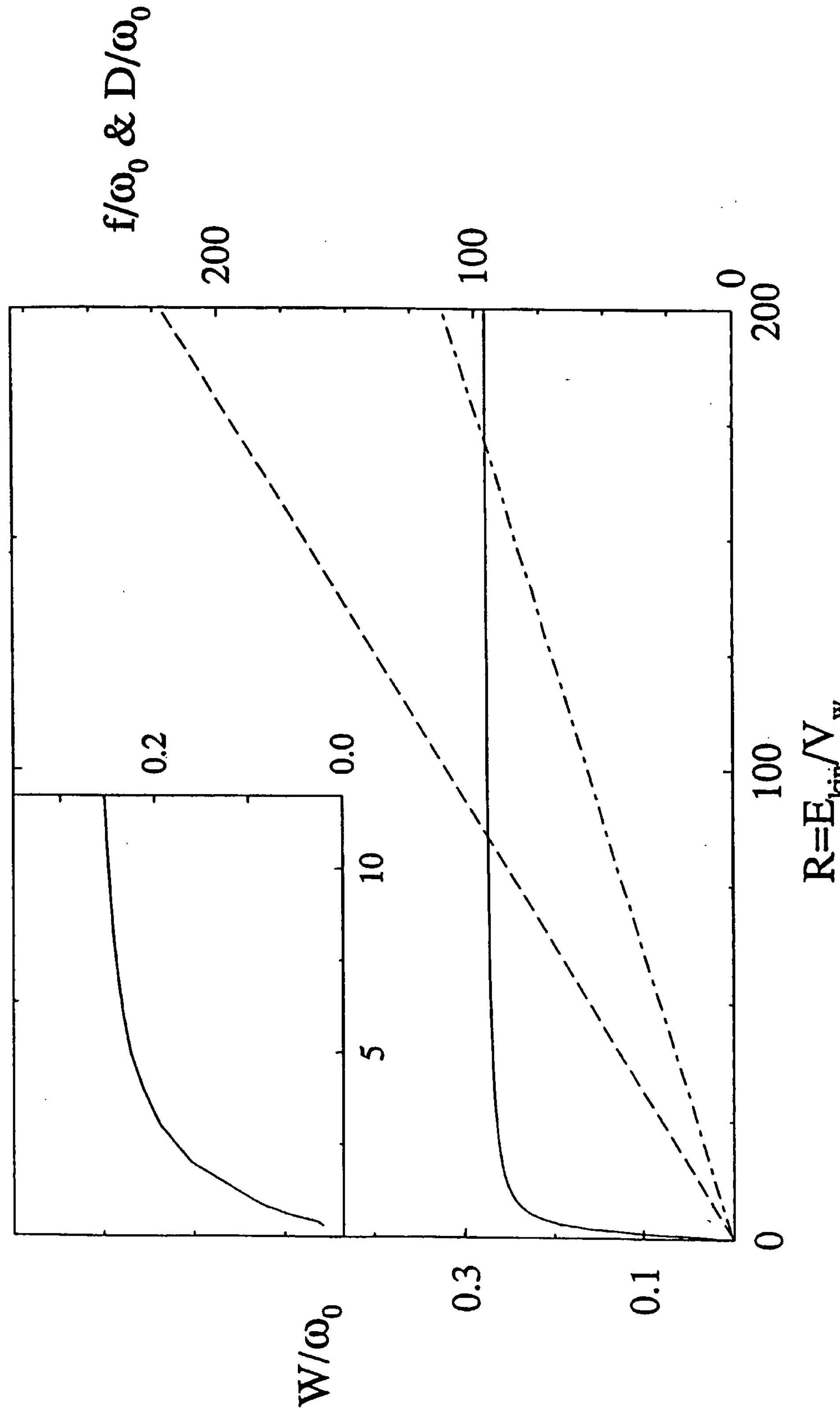


Figure 2a

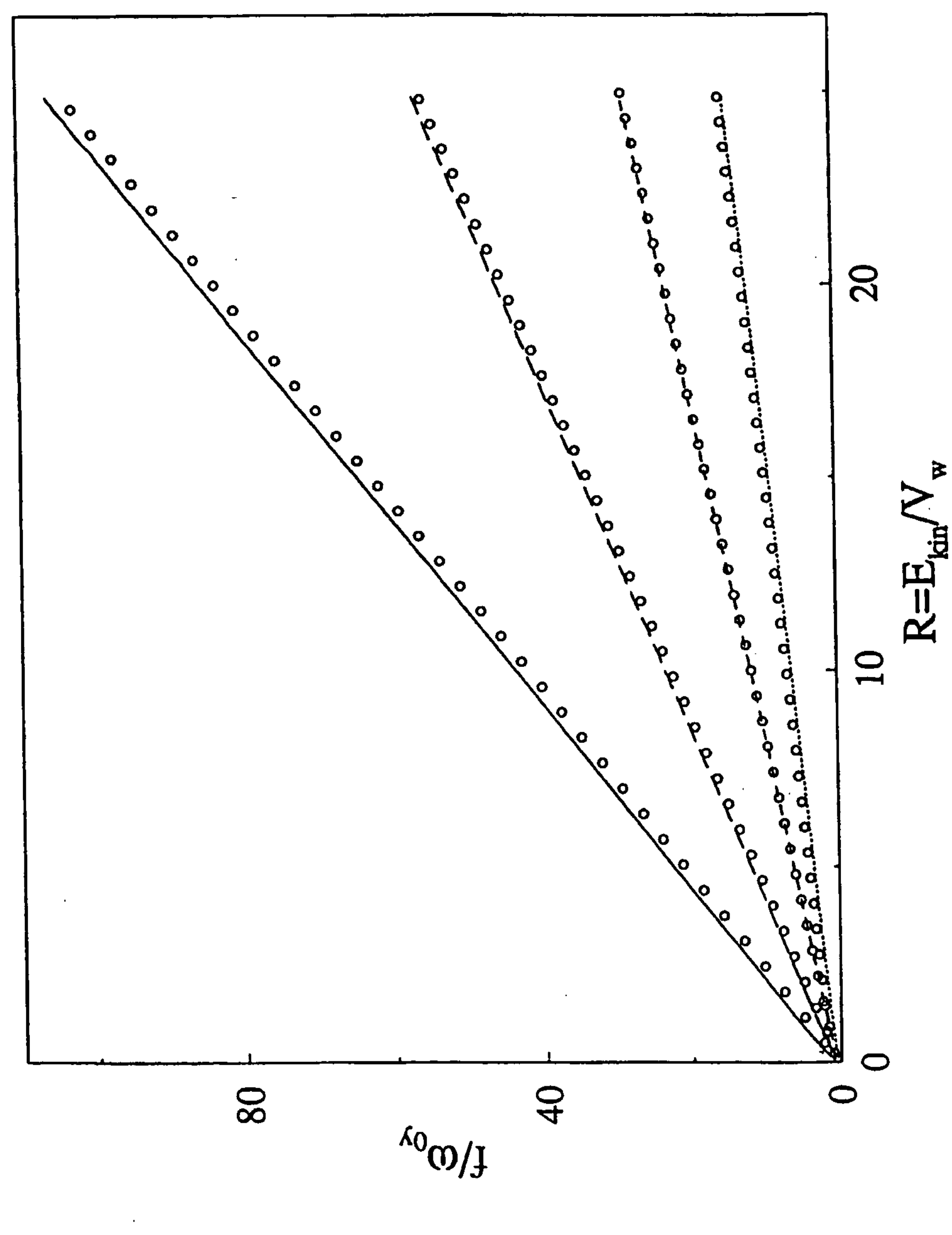


Figure 2b

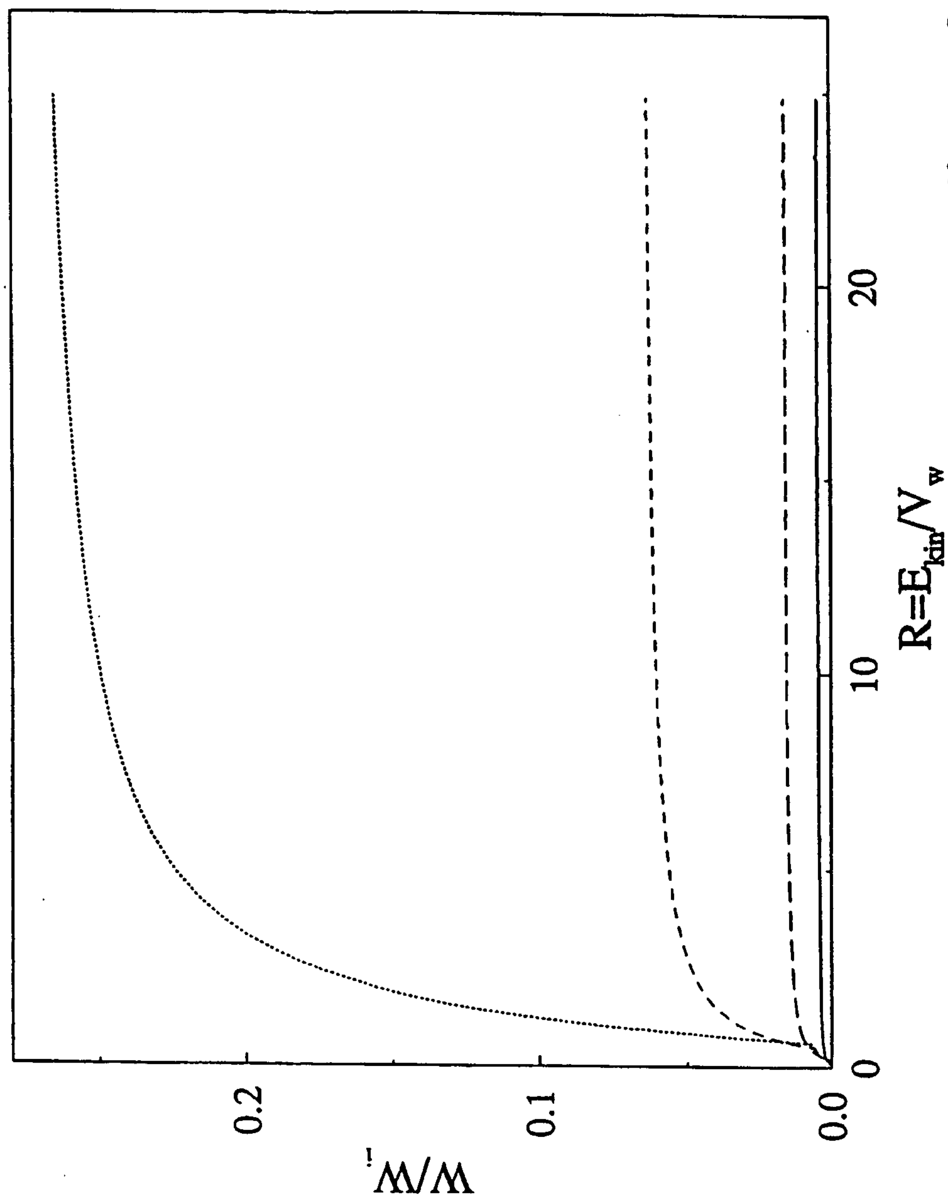


Figure 2c

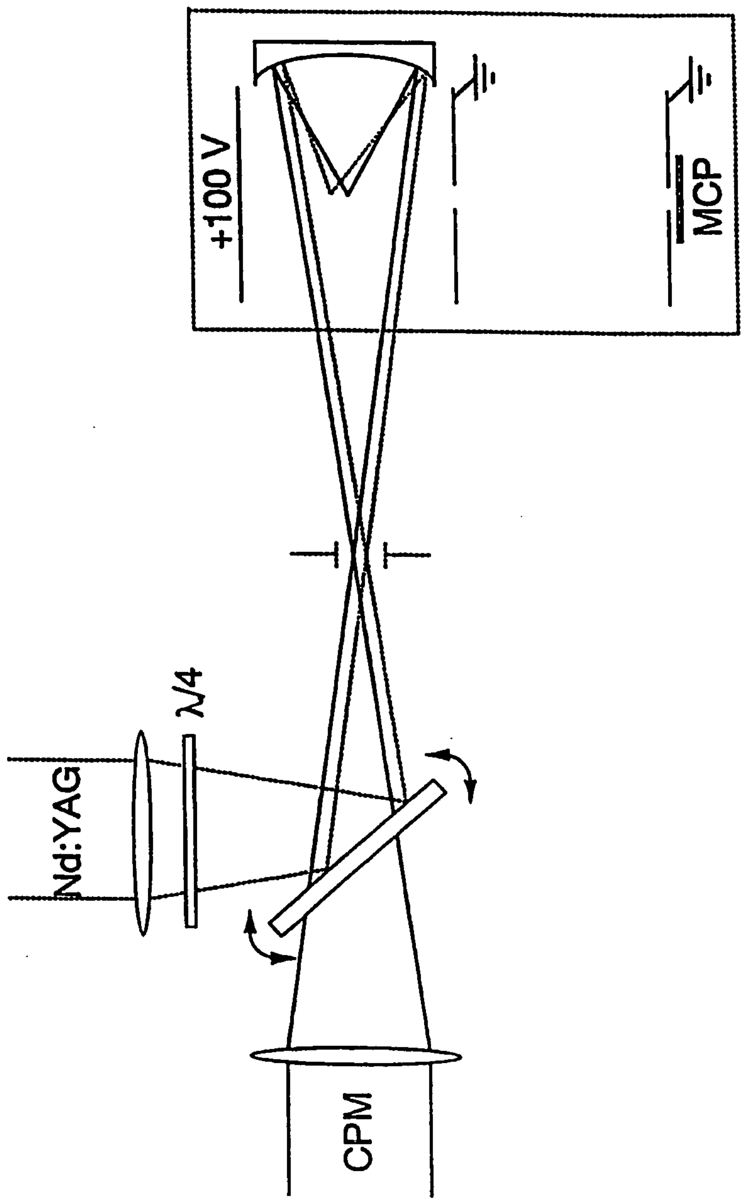
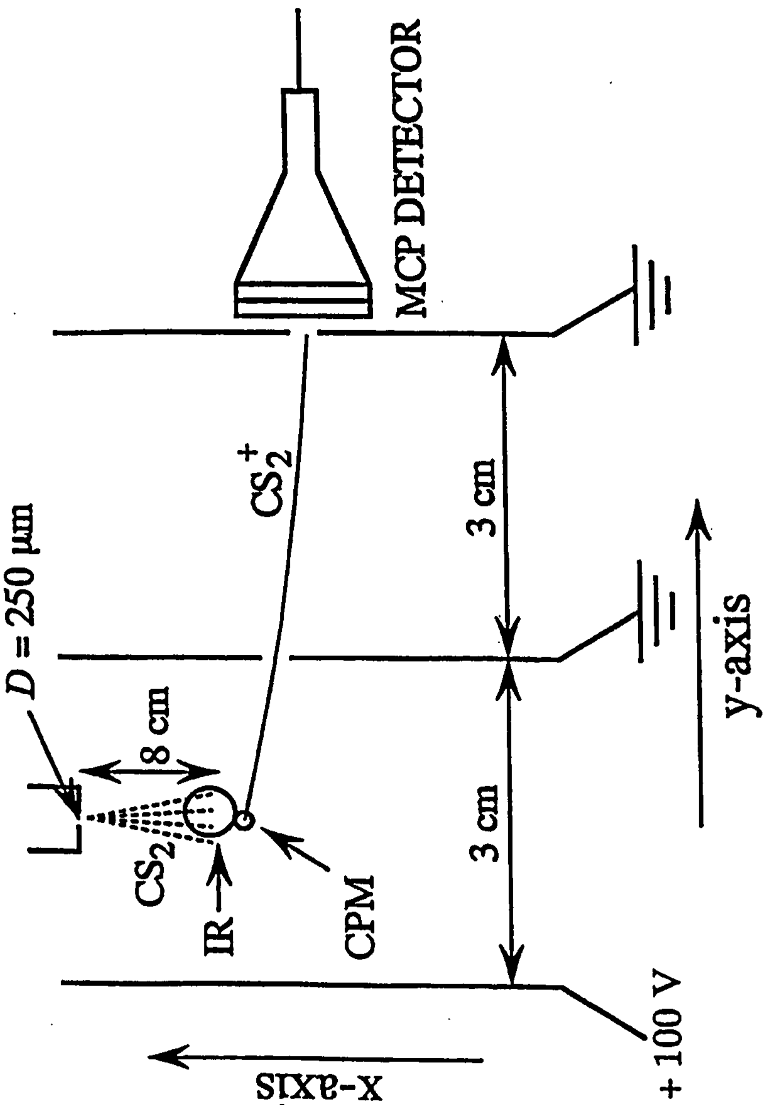


Figure 3a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CS2
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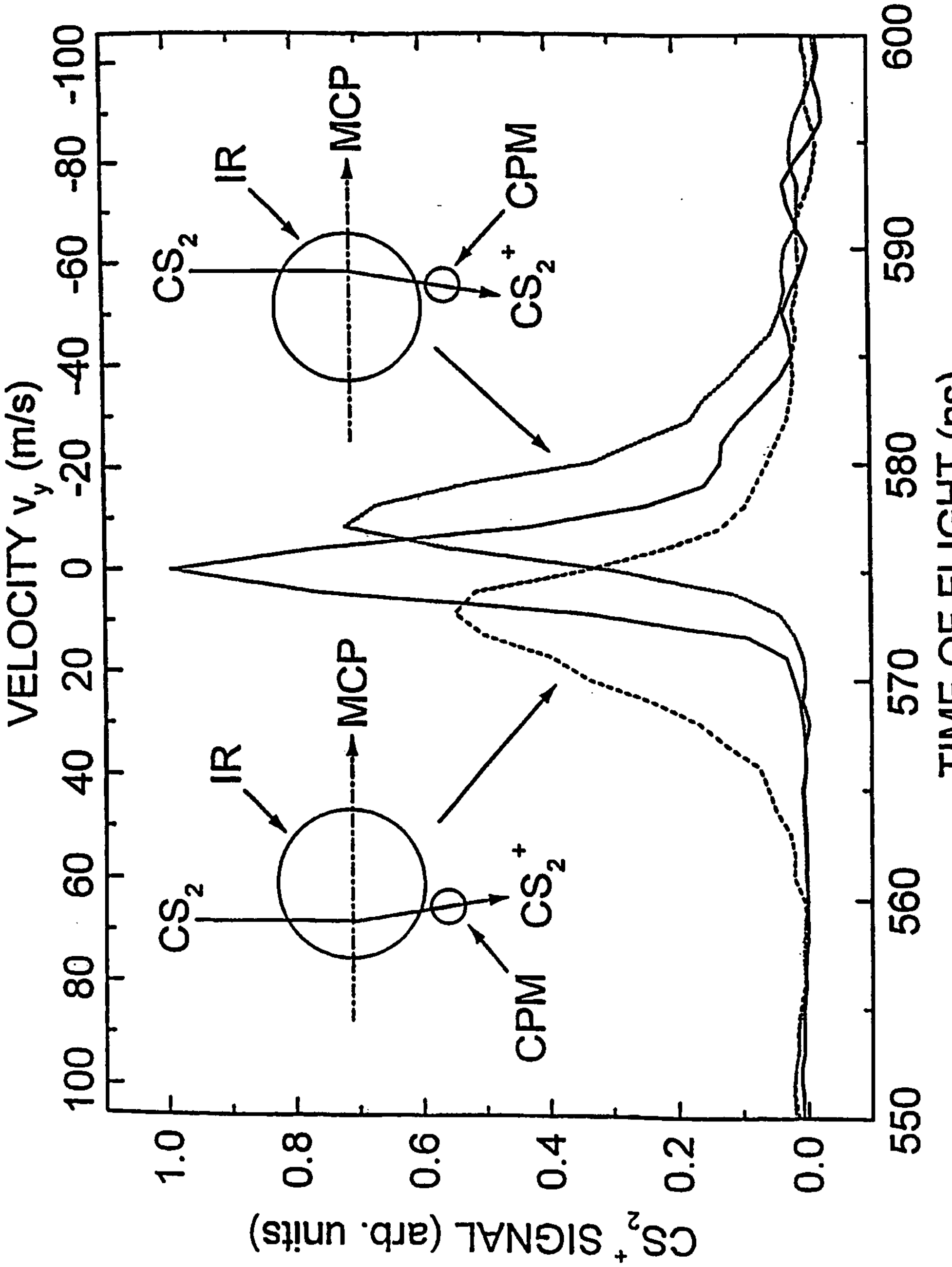
CPM
+ 100 V
x-axis
y-axis

Figure 3b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TIME OF FLIGHT (ns)
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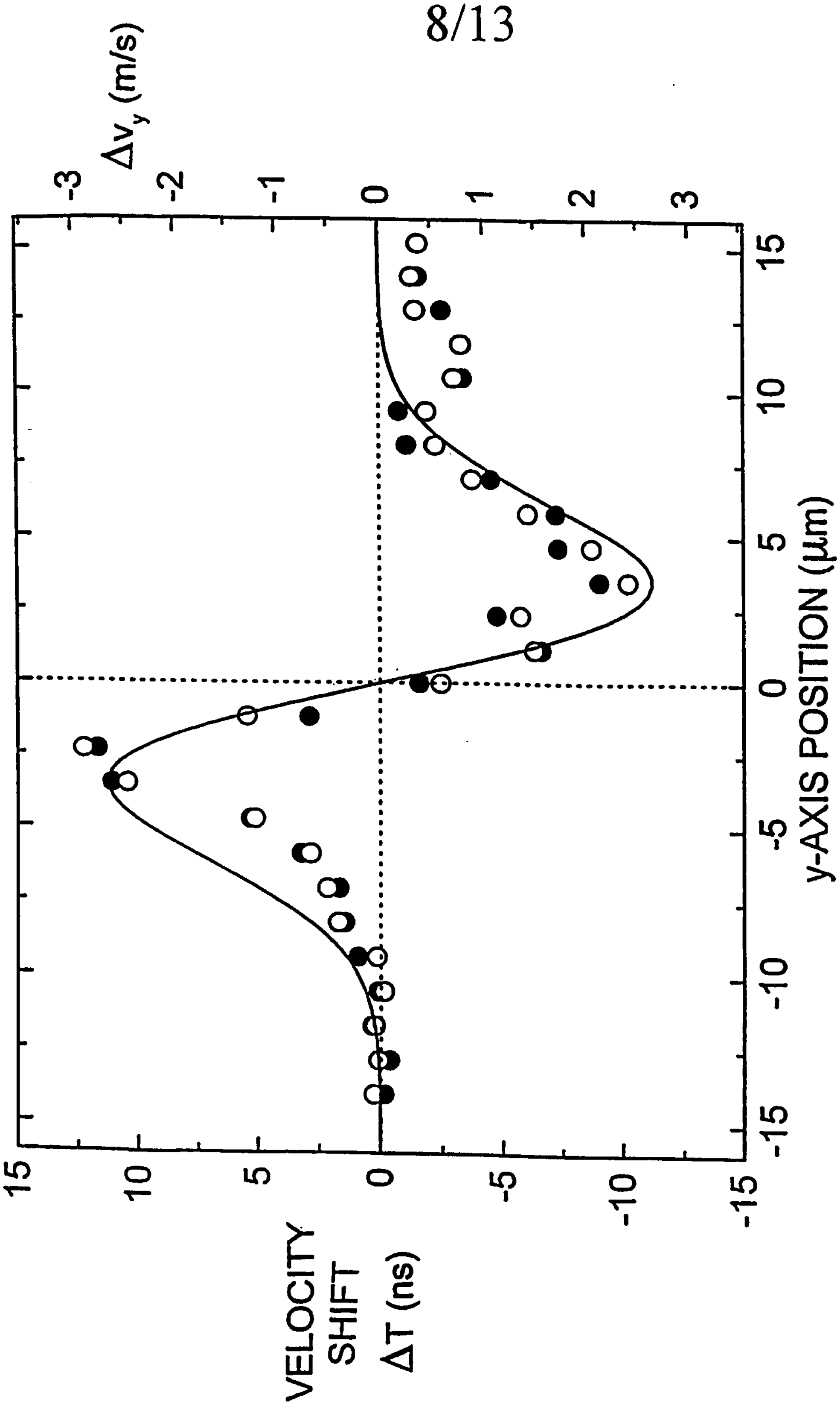
CS_2
IR
MCP
CPM
CS_2^+

Figure 4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ΔT (ns)
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Figure 5

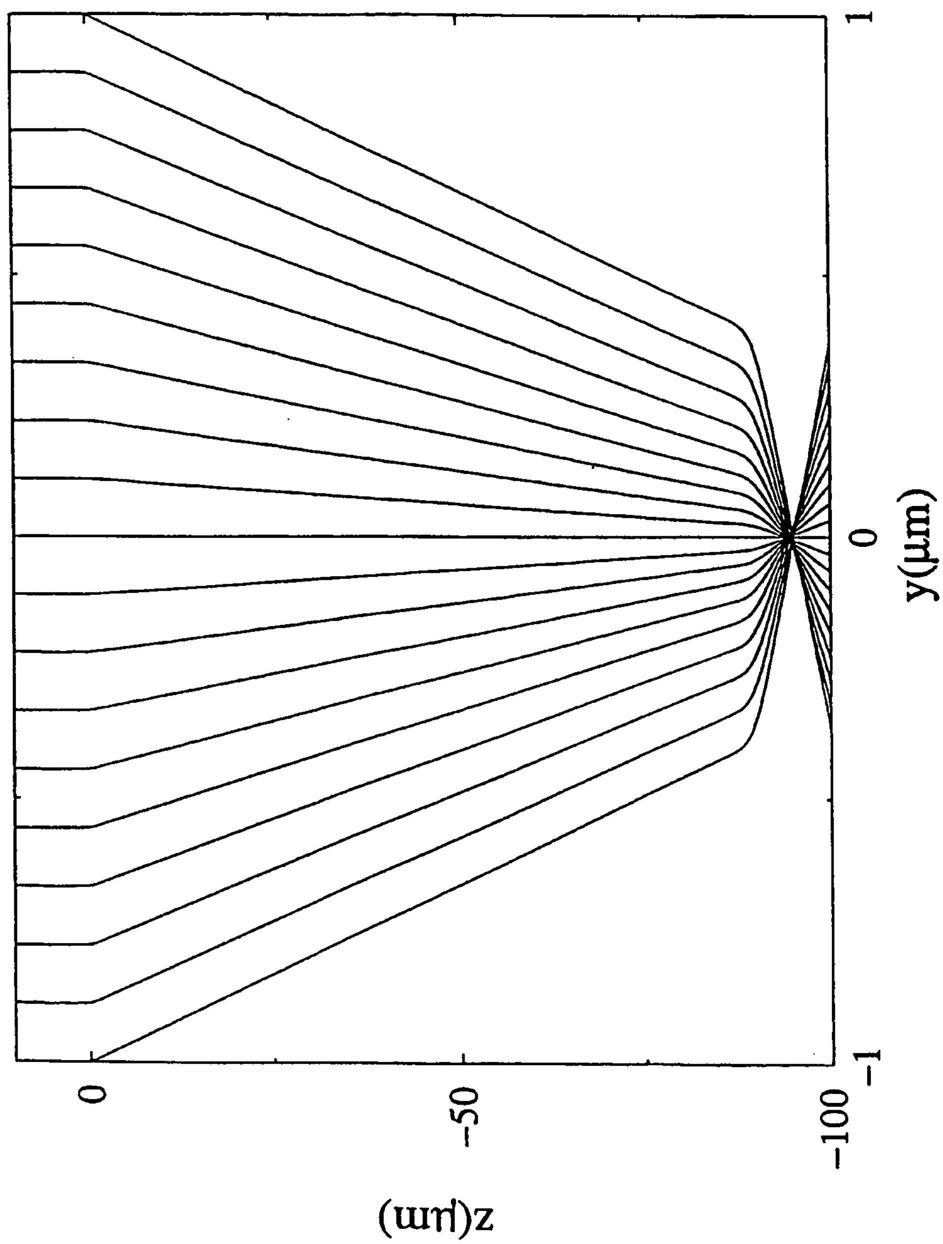

Figure 6

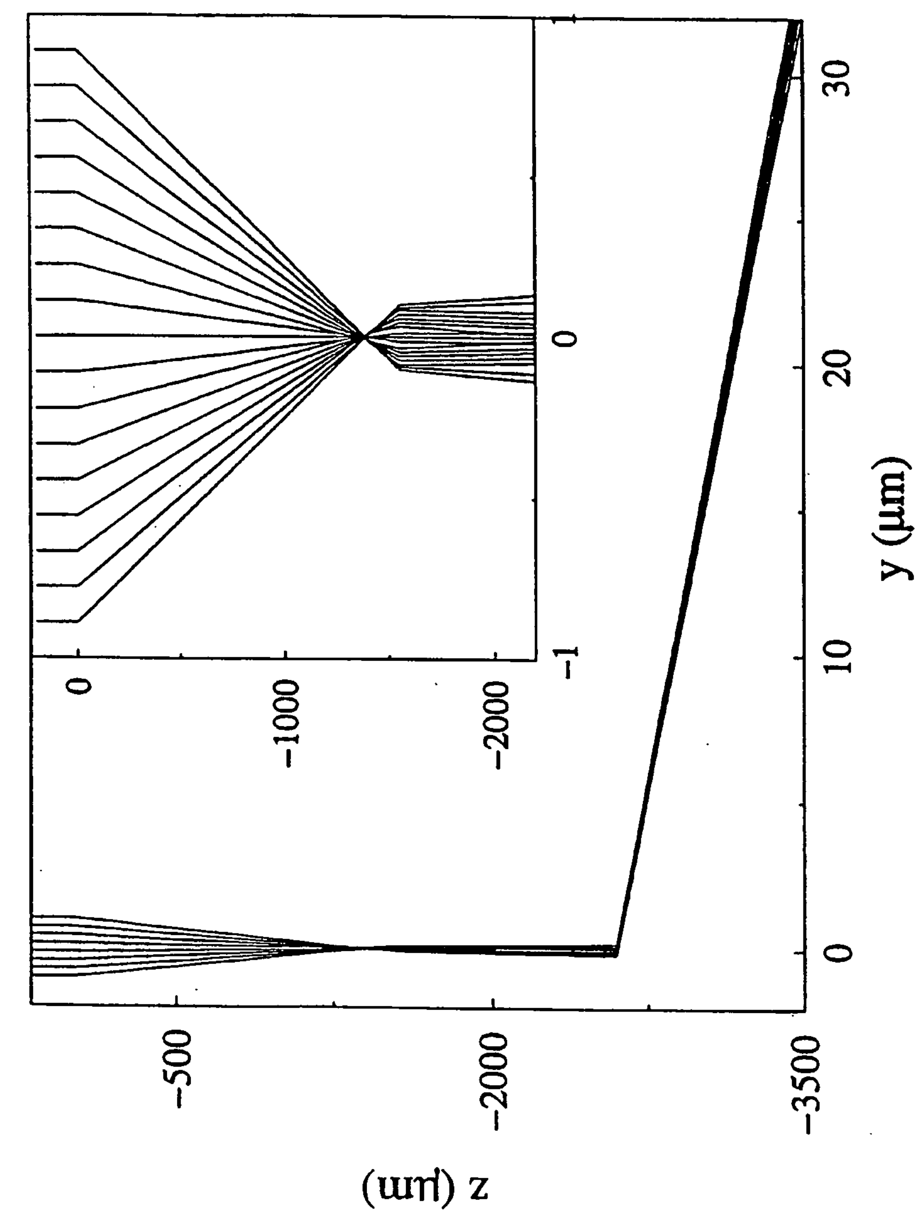

Figure 7

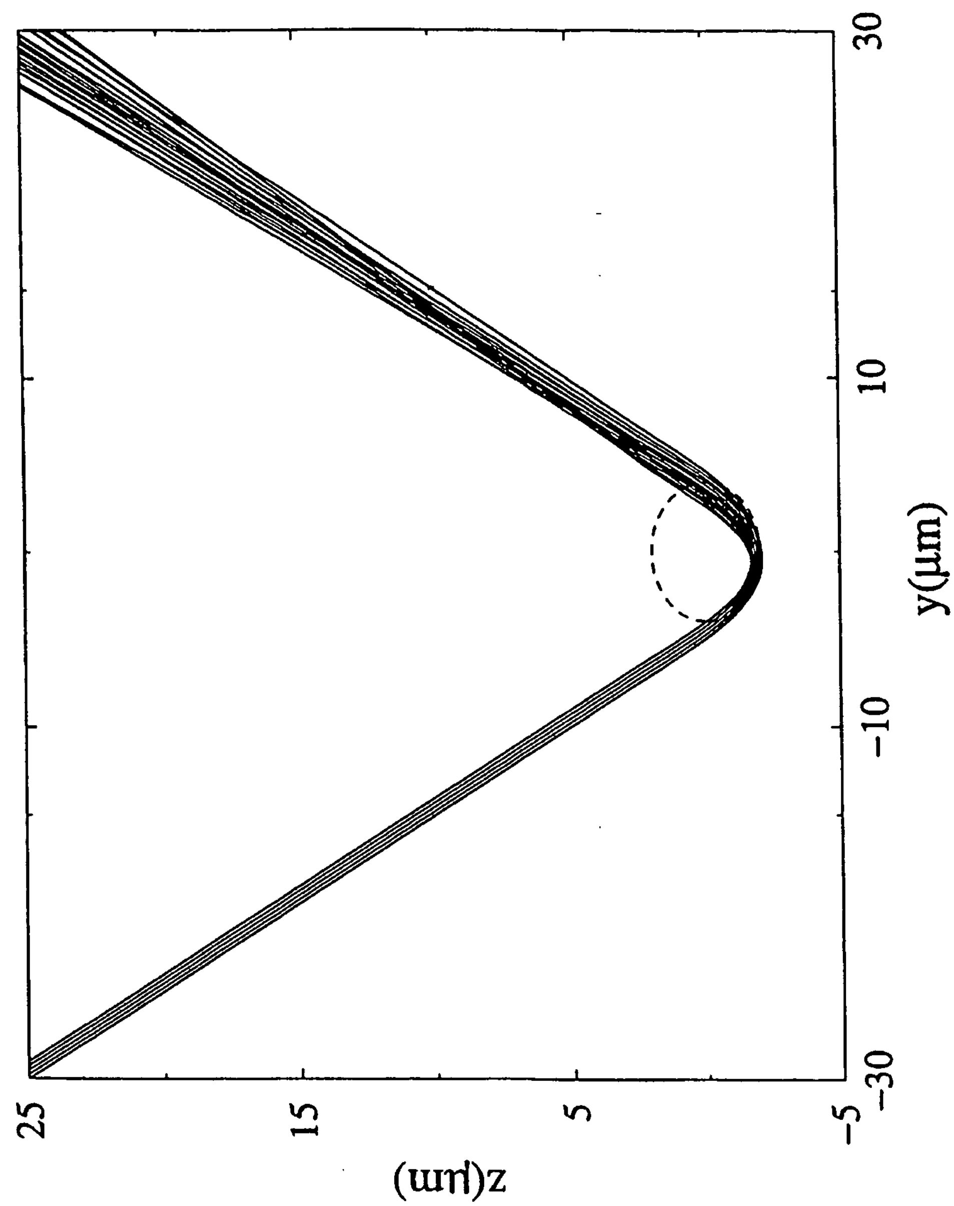

Figure 8

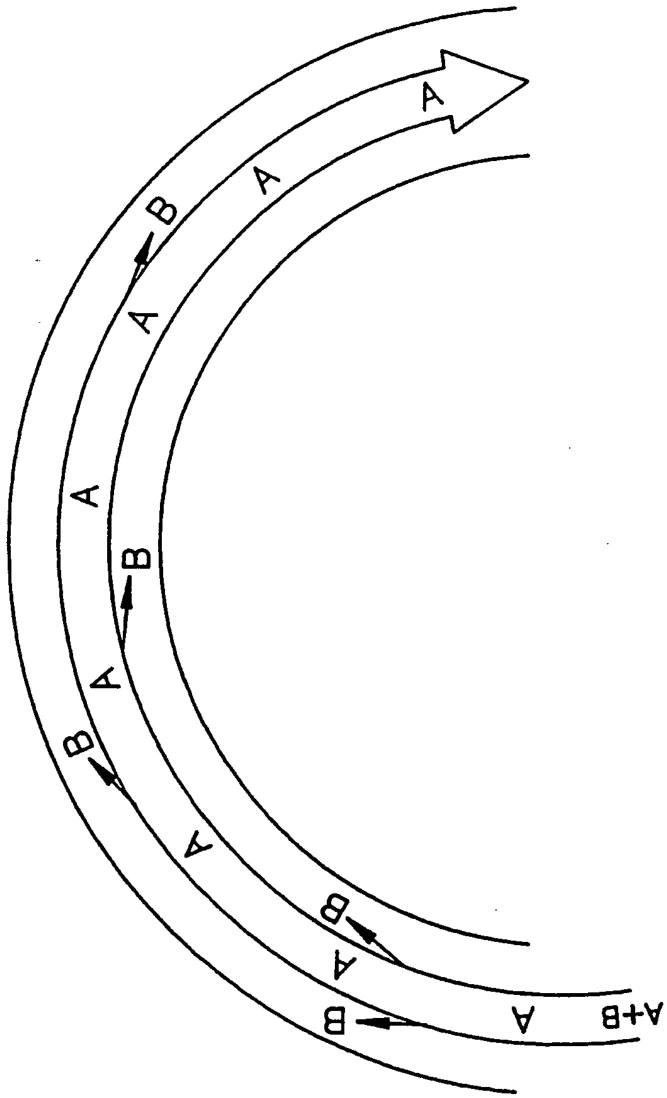

Figure 9a

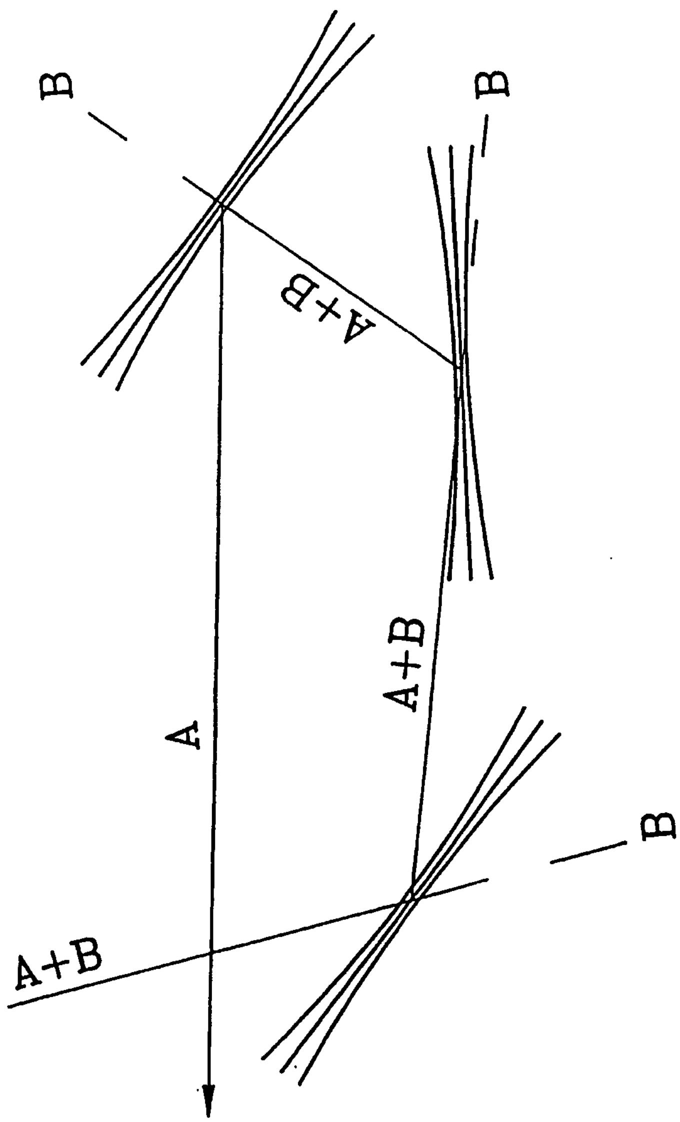

Figure 9b

INTERNATIONAL SEARCH REPORT

Inte. .ional Application No
PCT/CA 97/00911

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 H05H3/04

According to International Patent Classification(IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 H05H

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,X	STAPELFELDT H ET AL: "Deflection of neutral molecules using the nonresonant dipole force" PHYSICAL REVIEW LETTERS, 13 OCT. 1997, APS, USA, vol. 79, no. 15, ISSN 0031-9007, pages 2787-2790, XP002059271 see page 2787, paragraph 1 - page 2788, right-hand column, paragraph 1 see page 2790, left-hand column, paragraph 1 - paragraph 4 ---	1-7, 10-14

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publicationdate of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

1

Date of the actual completion of theinternational search	Date of mailing of the international search report
24 March 1998	06/04/1998
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer Capostagno, E

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INTERNATIONAL SEARCH REPORT

Inte. ,onal Application No
PCT/CA 97/00911

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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A	LINSKENS A F ET AL: "Alignment and orientation of nonpolar molecules utilizing the laser-induced AC-Stark effect" JOURNAL OF CHEMICAL PHYSICS, 1 DEC. 1994, USA, vol. 101, no. 11, ISSN 0021-9606, pages 9384-9394, XP002059273 see abstract see page 9389, left-hand column, paragraph 2 - right-hand column, paragraph 1 see figure 4 ---	1,4,6,7
A	GB 1 341 683 A (WESTERN ELECTRIC CO) 25 December 1973 see page 1, line 27 - line 40 see page 4, line 7 - line 43 ---	2,18,24, 30,34
A	US 5 170 890 A (WILSON STEVEN D ET AL) 15 December 1992 see the whole document ---	6,21
A	EP 0 490 697 A (JAPAN RES DEV CORP) 17 June 1992 see column 3, line 40 - column 4, line 18 -----	17,20,29

1

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

Information on patent family members

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PCT/CA 97/00911

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